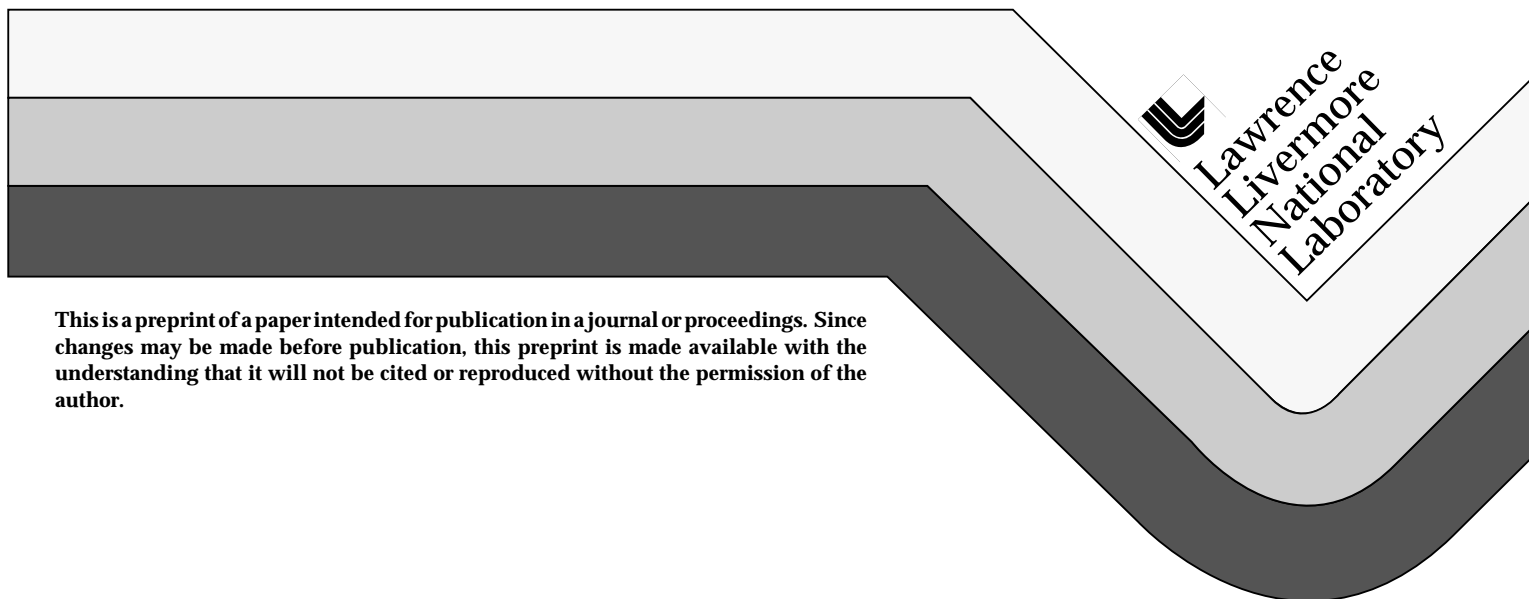


Polyimide Capsules May Hold High Pressure DT Fuel Without Cryogenic Support for the National Ignition Facility Indirect-Drive Targets

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POLYIMIDE CAPSULES MAY HOLD HIGH PRESSURE DT FUEL WITHOUT CRYOGENIC SUPPORT FOR THE NATIONAL IGNITION FACILITY INDIRECT-DRIVE TARGETS

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ABSTRACT

New target designs for the Omega upgrade laser and ignition targets in the National Ignition Facility (NIF) require thick (80 - 100 μm) cryogenic fuel layers. The Omega upgrade target will require cryogenic handling after initial fill because of the high fill pressures and the thin capsule walls. For the NIF indirectly driven targets, a larger capsule size and new materials offer hope that they can be built, filled and stored in a manner similar to the targets used in the Nova facility without requiring cryogenic handling.

I INTRODUCTION

The design of both direct-drive targets for the Omega upgrade laser facility and indirect-drive targets for the NIF facility require cryogenic hydrogen fuel layers with thickness in the range of 80 – 100 μm .

The Omega upgrade target designs require a very thin¹ (a few microns) capsule wall and a solid fuel layer thickness of 100 μm in a capsule of 1 mm ID. The calculated fill pressure needed for this target is 1100 atms or about 100 MPa. The only viable fabrication technique for this design will require cryogenic handling from the initial fill through storage, transfer, and fielding. The capsules will be diffusion filled and subsequently cooled to cryogenic temperatures before they are removed from the fill station. A transfer cryostat will then be used to retrieve the filled capsules from the high pressure fill cell and load them into a target storage and layering cryostat, where they will remain until needed in the target chamber. A transport cryostat will then take one capsule at a time to the target fielding cryostat in the Omega tar-

get chamber. This extensive use of a cryogenic environment will make the fielding of Omega's cryogenic targets cumbersome as well as expensive because of the time and personnel required to operate this complex cryogenic system.

The 2 mm diameter target capsules intended for indirect drive in the NIF with an 80 μm fuel layer thickness, which is roughly 20% thinner than the fuel layers for the Omega targets, makes the DT gas fill density about two to three times lower at 300 K for the NIF targets. The capsule wall thickness of 160 μm specified for the NIF targets is also much greater than the wall thickness stipulated for the Omega direct-drive design.

These differences in the design requirements of the NIF indirect-drive targets versus the Omega upgrade direct-drive targets may allow LLNL to construct capsules using materials capable of supporting sufficient fill pressure at room temperature to satisfy the requirements for the NIF indirect-drive targets. This capability will greatly reduce the cryogenic handling requirements and the cost of the target capsules for the NIF facility. Cryogenic temperatures will still be required for fuel layering and fielding, but that is much easier to achieve.

II FUEL FILL PRESSURE FOR NIF CAPSULES

The specifications² for an NIF ignition target with a CH_x ablator (the point design) call for a capsule with an ID of 0.95 mm, with a DT fuel layer thickness of 80 μm and a capsule wall thickness of 160 μm as depicted in Figure 1. The residual gas density of 0.3 mg./cc required by this

design determines the operating temperature (18.3 Kelvin) of the capsule.

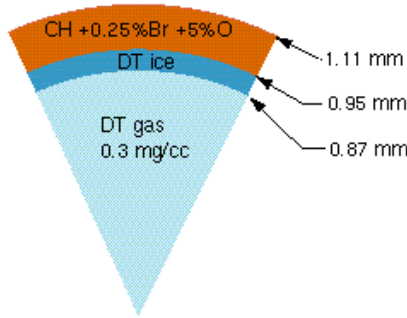


Figure 1. Sketch of cryogenic point design target for the NIF. The capsule has a DT ice layer 80 μm thick which requires a room temperature fill pressure of ~ 361 atm.

The gas fill density required to obtain the estimated fuel mass in the point design target with a capsule volume $V_c \approx 3.59 \text{ mm}^3$ is $\rho_{fill} \approx 60.5 (\text{mg/cc})$. This fill density is equivalent to a fill pressure² of $P_{fill} \approx 360 \text{ atm}$. The fill pressure required to obtain several different solid layer thickness is plotted in Figure 2 for D-T fill gas. The room temperature fill pressure decreases dramatically as the capsule diameter increases from 500 μm to 2 mm due to the decreasing surface to volume ratio.

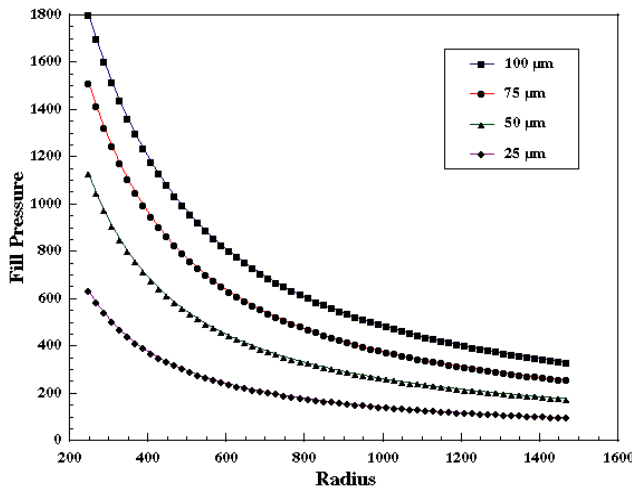


Figure 2. Required fill pressure (atm.) at 300K calculated vs. radius to produce a D-T solid layer at 18.3K

These lower fill densities may make it possible to obtain reasonably thick solid layers in diffusion filled capsules of large diameter without the requirement of cryogenic temperatures during fill and/or storage to prevent explosive self-destruction from overstress.

III CAPSULE STRESS CALCULATION AND REQUIRED MATERIAL TENSILE STRENGTH

The membrane stress generated on a thick walled (mean radius / thickness < 10) spherical pressure vessel by an internal pressure P_i is calculated and compared to the tensile strength of various common capsule materials.

On the diagram in Figure 3a is the outer radius, and b is the inner radius. The radial stress σ_r , reaches a maximum when $r=b$. The circumferential stress σ_c , and is The capsule material is assumed homogenous.,

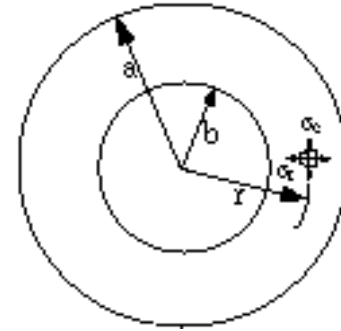


Figure 3. Reference diagram for stress calculations. The outer and inner radius are respectively a and b . The circumferential and the radial stresses are respectively σ_r and σ_c

The circumferential stress σ_c for a thick shell is given by the following equation assuming that the circumferential stresses are symmetric:

$$\sigma_c = \frac{P_i b^3}{2r^3} \cdot \frac{a^3 + 2r^3}{a^3 - b^3}$$

The equation for radial stress is:

$$\sigma_r = \frac{-P_i b^3}{r^3} \cdot \frac{a^3 - r^3}{a^3 - b^3}$$

The total compound stress is calculated from the sum of the absolute values of the radial and circumferential stresses as follows:

$$\sigma_{total} = |\sigma_r| + |\sigma_c|$$

The maximum stress is reached when $r=b$, at the inner capsule surface. A plot of the maximum compound stress for different fill pressures as a function of aspect ratio (*thickness/radius*) is shown in Figure 4.

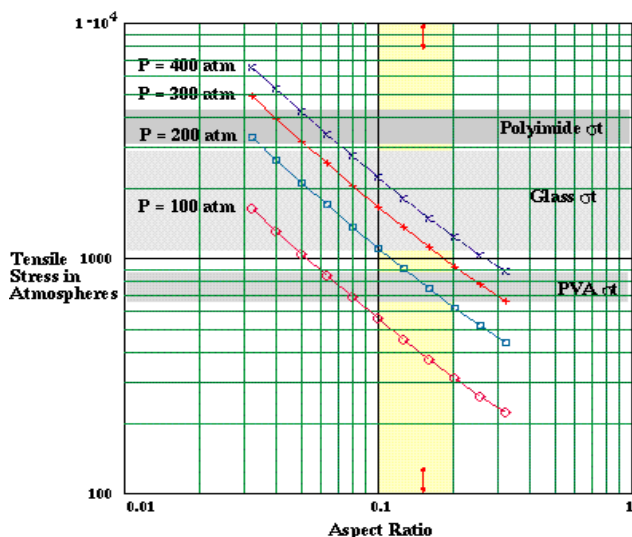


Figure 4. Calculated maximum compound stress for different fill pressures versus capsule aspect ratio. The tensile strength of polyimide, glass, and polyvinyl-alcohol (PVA) is also plotted for comparison. The little double arrow marker indicates where the aspect ratio of the NIF point design target falls in the graph.

The tensile strength of different capsule materials has been plotted for comparison. For the fill pressure of 360 atmospheres required for the NIF point design target, both polyimide and glass may have sufficient strength to support the room temperature stress generated by the fill. Glasses are not considered desirable candidates because of their high oxygen² content.

IV CHARACTERISTICS OF POLYIMIDES APPLICABLE TO TARGET REQUIREMENTS

A. Polyimide Tensile Strength

Table 1 lists a number of polyimide formulations in order of decreasing tensile strength⁴. The polyimide used to manufacture the Laser Entrance Holes (LEH) windows is the Hitachi L-100. Its tensile strength⁵ has been measured by Ed Hsieh on thin films <1.0 μm using a bi-axial stretching technique. The measured strength of the films compared well with the strength quoted by Hitachi. For comparison

the tensile strength of some common capsule materials are also listed.

TABLE 1. Tensile strength comparison

Product name or formulation	Tensile Strength atm.	Comments
BPDA / PPD	3350- 4180	can be vapor deposited
6FCDA / TFMOB	4050	
PMDA / TFMOB	3730	
PMDA / TFMB	3690	
PMDA / TFEOb	3290	
BPDA / TFMOB	3120	
Hitachi L-100	3100	Used on LEH windows
Glass	990 - 1970	
PVA	590 - 990	Permeability barrier
Formvar	610	Capsule support film
Polystyrene	800	Capsule mandrels

As can be readily determined from the table, many polyimide formulations have tensile strengths that range from 1.5 to 4 times larger than glass.

Polyimides are synthesized from precursor dianhydrides and diamines.

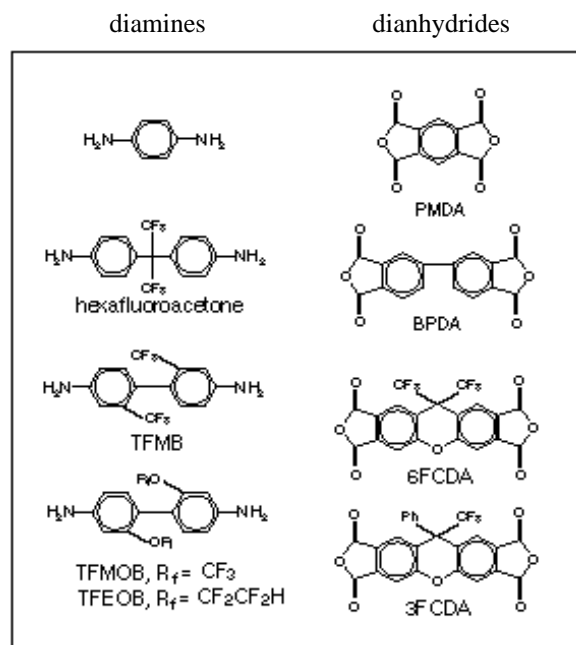


Figure 5. Chemical composition of a small sample of diamines and dianhydrides used to make polyimides with different material characteristics.

These precursors, when combined in the proper stoichiometric ratios, react to form poly(amic acids). Since

there are many possible permutations of variations of these two components, see Figure 5, the material strength and permeability may be tailored^{4,6} for a given application

The tensile strength of polyimide depends not only on its particular chemical structure, but also on the imidization process itself. The imidization step is essentially a curing process that changes the chemical structure of the poly(amic acids) into polyimide.

This process is generally accomplished by elevating the temperature of the deposited poly(amic acids) in a prescribed sequence of steps. First to evaporate residual solvent, and later at a higher temperature to activate the transformation of poly(amic acids) into polyimide. Like many thermally activated processes, this process is amenable to precise temperature control; thus the process dependence of the tensile strength can be controlled⁷.

Although the imidization process can also be accomplished chemically, it is not widely employed because of the problems associated with handling the reagents.

B. Polyimide permeability and fuel retention times

The hydrogen permeability of polyimides varies over several orders of magnitude, depending not only on the chemical composition of the particular polyimide, but also on whether the film was made using a solvent based deposition or vapor phase⁸ deposition.

Permeability coefficients measured using O₂ as a test gas for vapor phase deposited polyimide⁸ PPD-TCL were as much as 1/100 of the value measured for solvent based coatings of the same PPD-TCL material. If the particular polyimide formulation can be deposited using both methods, then the permeability may be adjusted without changing the chemical structure of the polymer, a potentially useful characteristic of polyimide fabrication.

The hydrogen permeability⁹ of some polyimides are listed in table 2. The permeability values for PVA and Polystyrene are also included for comparison.

TABLE 2. Permeability of Polyimides

Product name or formulation	Permeability x10 ⁻¹⁵ in (mole m) / (K m ² s Pa)	Comments
Clear PPD / TCL	0.000107 – O ₂ @ 300K	vapor dep. ¹⁰
Hazy PPD / TCL	0.000335 – O ₂ @ 300K	vapor dep. ¹⁰
PPD / TCL	0.0127 – O ₂ @ 300K	solvent dep. ¹⁰
TMAC / ODA	1.41 – H ₂ @ 373K	solvent dep. ¹¹
PMDA / ODA	0.08 – O ₂ @ 373K	solvent dep. ¹¹

TABLE 2. Permeability of Polyimides

Product name or formulation	Permeability x10 ⁻¹⁵ in (mole m) / (K m ² s Pa)	Comments
PMDA / ODA	8.00 – H ₂ @ 373K	solvent dep. ¹¹
BTDA / ODA	5.49 – H ₂ @ 373K	solvent dep. ¹¹
BTDA / mTrMPD	23.5 – H ₂ @ 310K	solvent dep. ¹¹
BPDA / mTrMPD	66.7 – H ₂ @ 310K	solvent dep. ¹¹
Hitachi L-100	n/a	solvent, LEH
PVA	0.000239 – H ₂ @ 300K	H ₂ O conc. Dep.
Kapton	0.596 – H ₂ @ 300K	solvent dep.
Polystyrene	18.8 – H ₂ @ 300K	Cap. Mandrels

The leak rate time constant from a thin wall spherical container is $\lambda = (kAR_0T)/tV$ where k is the material permeability, A is the capsule's mean surface area, R_0 is the universal gas constant, T is the temperature (Kelvin), t is the capsule's wall thickness, and V is the capsule's volume. Using the H₂ permeability of Kapton I obtain a fuel retention time half-life ($(\sqrt{2})/\lambda$) of ~7 hours for a 160 μ m thick, 2 mm ID capsule at room temperature -- a little short to accommodate the target assembly time for an NIF cryogenic target.

The permeability measurement for the PPD / TCL polyimide were done with O₂. Using the O₂:H₂ permeability ratio of the PMDA / ODA polyimide, I estimate the Hydrogen permeability of the PPD / TCL polyimide to be 100 times greater for H₂ than for O₂. The estimated Hydrogen permeability of PPD / TCL is about 5 times smaller than the permeability of Kapton. Scaling the fuel retention half-life obtained for Kapton by a factor of 5 would give a half-life of ~35 hours at room temperature.

The fuel retention time half-life of ~50 hours for a typical 500 μ m ID capsule with a 2 μ m thick PVA layer is sufficient to accommodate present day assembly requirements. Assuming that the NIF hohlraum target assembly time will be similar, the ~35 hours fuel retention half-life would be short, but still provide adequate assembly time.

For an ideal capsule, if it were manufactured from a single component, the required H₂ permeability for a ~100 hours fuel retention half-life would be approximately a twentieth of the permeability of Kapton film at room temperature. Vapor deposition of polyimides may provide the required lower permeability values.

C. Other advantages of using polyimides

There are several additional advantages of polyimides that have a direct impact on target fabrication technology.

Polyimides have excellent resistance to radiation induced damage. DuPont's VESPEL polyimide is often used for valve seat applications in valves intended for Tritium service. It is more resistant than most polymers, and does not produce corrosive gases (hydrogen chloride, hydrogen fluoride) like halogenated polymers.

Most polyimides are transparent to visible radiation and will not impede capsule surface or fuel layer characterization with optical methods. Other materials such as Beryllium which are potentially better capsule materials, are totally opaque.

Polyimides also have lower absorption of IR radiation than CH_x in the band needed for IR^{10} smoothing ($\lambda = 2.5 - 3.2 \mu\text{m}$) of DT layers, but to further reduce the IR absorption (see Figure 6) to an acceptably low level it will be necessary to replace the hydrogen with deuterium. The polyimide's low dielectric constant should allow efficient RF induced layer smoothing if required.

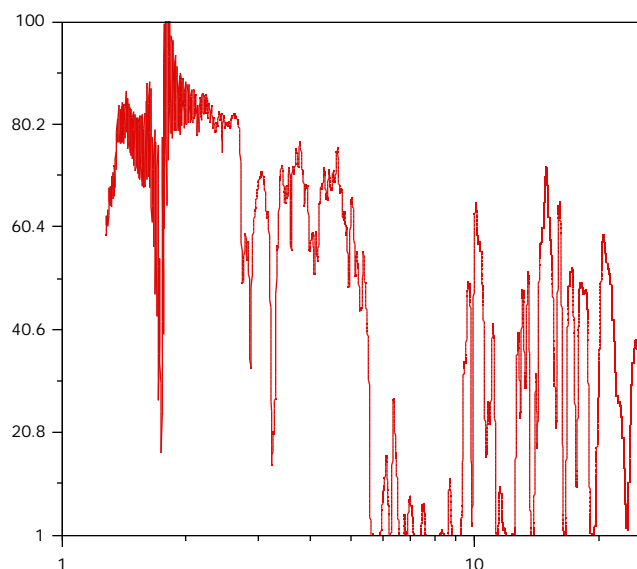


Figure 6. Kapton polyimide% absorption vs. wavelength (μm)

Their high temperature stability (break down temperatures often exceed 500°C) and strength at the higher temperatures are also highly desirable characteristics that will ease the capsule fill process.

IV CAPSULE FABRICATION ISSUES

There are several possibilities for making capsules out of polyimides. Solvent based poly(amic acids such as the Hitachi L-100 used to make hohlraum windows may be

used directly in drop towers¹¹, but the desired thick walls will require the use of external forces (acoustic) to center the bubble within the shell.

The liquid microencapsulation technique may also be applied directly to making shells from the solvent based material, but compatible liquids capable of allowing diffusion of the solvents from the formed shell as it dries are required. And the centering of the inner bubble on the shell may still be required, but it should be more straightforward to use acoustic centering in a liquid environment.

Vapor deposition⁸ of the polyimide onto a suitable substrate is one of the more promising techniques for the fabrication of target capsules. It is a proven technique for the deposition of thin films (it is used for conformal coatings in the fabrication of IC's).

Polyimides made via the vapor deposition method are often made by co-evaporating a binary combination of a diamine and a dianhydride which allows for many different polyimides to be made. However this technique relies on maintaining tight control on the stoichiometric ratio of the precursor monomers in the vapor phase, a sometimes difficult problem.

A different approach proposed¹² for vapor deposition that removes the need to keep the proper stoichiometric ratio relies on the possibility of making a monomer of the poly(amic- acid) that is then vaporized by sublimation and deposited directly on the substrate. With this technique, the deposition process should be simpler and more reliable with the possibility of obtaining much better surface finishes.

V PLASMA PHYSICS ISSUES: IMPACT OF POLYIMIDE ON CAPSULE IMPLOSION DYNAMICS

The mayor impact that polyimide based capsules have on the capsule implosion dynamics is due to the larger oxygen content of the polyimide material. The plasma deposited CH_x ablators presently used on target capsules contains approximately 5% oxygen², and the PVA permeability barrier contains 11% oxygen.

The chemical formula for the Hitachi PIQ-L-100 polyimide is $\text{C}_{22}\text{H}_{10}\text{N}_2\text{O}_4$ which has a 10.5 (atomic%) oxygen content. The polyimide with the highest measured tensile strength, BPDA / PPD (Biphenyldianhydride / 1,4 Phenylenediamine), has a formula $\text{C}_{22}\text{H}_{12}\text{N}_2\text{O}_4$ which results in 10.0 (atomic%) oxygen content.

The extra oxygen increases the x-ray absorption of the polyimide producing a narrower ablation front during

the implosion. This narrower ablation front induces higher peak acceleration at the ablation interface making it more susceptible to the formation of Raleigh-Taylor instabilities. As a consequence, the capsule's surface finish requirement may become more severe. Determination of this trade-off will have to wait until calculations¹³ of capsule implosion dynamics are performed using ablators with the appropriate chemical structure.

VI CONCLUDING REMARKS

Target capsules manufactured from polyimides can have a large impact on the design and cost of the capsule DT fill facility and the cryogenic target fielding and fuel layering system for the NIF.

The high tensile strength of polyimide capsules should support the required high fuel pressure at room temperature. No cryogenic support would be required following the fill process, thus greatly reducing the complexity of the fill system as well as reducing the quantity of DT stock required to fill NIF targets.

The good radiation resistance of polyimides would allow an inventory of filled capsules to be kept further reducing the availability requirements of the fill facility while de-coupling the same from the target layering and fielding process.

Low IR absorption in the region of 2.5 - 3.5 μm as well as low dielectric constant and low losses at RF frequencies would allow enhanced layer smoothing with either technique should that become necessary for ignition targets.

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REFERENCES

1. C. P. Verdon, Bull. Am. Phys. Soc. 38, 2010 (1993).
2. John Lindl, "Development of the indirect drive approach to inertial confinement fusion and the target physics basis for ignition and gain," Physics of Plasmas Vol. 2 No. 11, (November 1995), p 3968.
3. Roark and Young, *Formulas for Stress and Strain*, 5th Edition, p 503, McGraw-Hill Book Company, New York, 1975.
4. Andrew E. Feiring, Brian C. Auman, and Edward R. Wonchoba, "Synthesis and Properties of Fluorinated Polyimides from Novel 2,2'-Bis(fluoroalkoxy)benzidines" Macromolecules 1993, 26, 2779-2784.
5. Ed Hsieh and others, "The Strength and Other Information on Hitachi L-100 Polyimide Films," 10th Target Fabrication Specialists' Meeting, LA-UR-95-2938.
6. "Aromatic Copolyimide Membranes for High Temperature Gas Separation: H₂/CH₄, H₂/N₂, and O₂/N₂" Journal of Applied Polymer Sciences, Vol 41, 1261-70 (1990).
7. The polyimide windows manufactured by LUXEL for the hohlraum's laser entrance holes are made from polyimide film. According to Dr. Forbes Powell from LUXEL, the polyimide film used in the manufacture of the LEH windows, is spin cast and thermally cured. The curing process is carried out in a vacuum oven with a precisely controlled thermal ramping and soaking periods to insure repeatable tensile strength for the film.
8. R. M. Ikeda, R. J. Angelo, F. P. Boettcher, R. N. Blomberg, and M. R. Samuels "Polymerization from the Vapor Phase. I. Poly(p-phenyleneterephthalamide) Gas Barrier Coatings," Journal of Applied Polymer Science, Vol 25, 1391-1405(1980).
9. H. C. W. M. Buys, A. Van Elven, A. E. Jensen, and A. H. A. Tinnemans, "Aromatic Copolyimide Membranes for High Temperature Gas Separation: H₂/CH₄, H₂/N₂, O₂/N₂," Journal of Applied Polymer Science, Vol 41, 1261-70 (1990).
10. G. W. Collins, and others, "Infrared redistribution of D₂ and HD layers for inertial confinement fusion," J. Vac. Sci. Technol. A 14(5), Sep/Oct 1996. p 2897.
11. J. H. Campbell, and others, "Preparation and properties of hollow glass microspheres for use in laser fusion experiments," UCRL-53516.
12. Private communication with Don Brandom.
13. Private communication with S. Haan